Chemical selection of emission state configuration in a quantum-light emitter

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Los Alamos National Laboratory researchers and their collaborators have found a new way to control quantum-light emitters using so-called zigzag nanotube structures. Their work is published in the latest edition of *Nature Chemistry*.

“Our effort at Los Alamos has been advancing the development of chemically controlled defects in carbon nanotubes as room-temperature single photon emitters. These are of significant interest for enabling new optical approaches to quantum information processing,” said Stephen Doorn, one of the authors, and a scientist with the Los Alamos Center for Integrated Nanotechnologies (CINT). “Our latest work aims to address how to control and narrow the emission wavelength range through control of functionalization chemistry.”

Previous work from the group at CINT has shown that covalent chemical functionalization can lead to multiple binding configurations of the defects, or dopant molecules. These configurations in turn lead to a broad range of possible defect-state photoluminescence emission wavelengths. However, applications of this research require a significantly narrower wavelength range in the emission response, in order to provide a high degree of uniformity.

To take this next step, Lab researchers from CINT and the Center for Nonlinear Studies (CNLS) teamed up with colleagues from the National Institute of Standards and Technology (NIST), North Dakota State University and Japan’s Advanced Industrial Science and Technology (AIST). They showed that introducing defect-states into zigzag carbon nanotubes can narrow the emission wavelength range by a factor of 4-6. The narrowed response results from the unique symmetry of the zigzag nanotube’s physical structure.

Using quantum chemical theory modeling of the response, the researchers gained further insight into the origin of the spectral narrowing. The theory results provided evidence that only a particular type of chemical binding–binding to so-called “ortho” binding sites–results from the functionalization chemistry used by the team. This is the first demonstration of such binding selectivity, and it provides a previously missing atomistic-level understanding of carbon nanotube reaction chemistry.

“This latter discovery is an especially significant advance in our understanding of the reaction mechanisms in carbon nanotube chemistry,” Doorn said.

Publication: A. Saha, B. J. Gifford, X. He, G. Ao, M. Zheng, H. Kataura, H. Htoon, S. Kilina, S. Tretiak, S. K. Doorn, “Narrow-Band Single-Photon Emission through Selective

Aryl Functionalization of Zigzag Carbon Nanotubes", *Nature Chemistry*, 2018, *DOI: 10.1038/s41557-018-0126-4*.

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